Field-induced transition between magnetically disordered and ordered phases in underdoped $La_{2-x}Sr_xCuO_4$

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Abstract

We report the observation of a magnetic-field-induced transition between magnetically disordered and ordered phases in slightly under-doped $La_{2-x}Sr_xCuO_4$ with $x = 0.144$. Static incommensurate spin-density-wave order is induced above a critical field of about 3 T, as measured by elastic neutron scattering. Our results allow us to constrain the location of a quantum critical point on the phase diagram.

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The interplay between magnetic order and superconducting order is one of the most fascinating and important subjects in the study of high-T_c cuprate superconductors.[1,2,3,4] Magnetic order or fluctuations are found in almost every member of this large family of materials, in both hole-doped and electron-doped superconductors, and in the entire doping range studied, from undoped antiferromagnetic insulators to the overdoped superconductors. At first, the magnetism in different cuprates appears to have different manifestations: incommensurate static or dynamic spin-density-waves in $La_{2-x}Sr_xCuO_4$,[1] a resonance peak at the antiferromagnetic zone center in $YBa_2Cu_3O_{6+x}$,[5,6,7] and $Bi_2Sr_2CaCu_2O_8$,[8] and 3D antiferromagnetic order in electron-doped $Nd_{2-x}Ce_xCuO_4$ and $Pr_{1-x}LaCe_xCuO_4$.[9,10] However, recent experiments suggest a more unified picture, at least for the hole-doped materials. It is found that characteristic features of both spin-density correlations and the resonance exist in $La_{2-x}Sr_xCuO_4$,[11] $La_{2-x}Ba_xCuO_4$,[12] and $YBa_2Cu_3O_{6+x}$.[13,14,15] In addition, high-energy neutron scattering studies show that the magnetic fluctuations have similar dispersion in $La_{2-x}Ba_xCuO_4$,[12] optimally-doped $La_{2-x}Sr_xCuO_4$[16] and $YBa_2Cu_3O_{6+x}$.[17,18] The importance of these results is that $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$ have very different superconducting T_c's, crystal structures, and other properties. Therefore, the This universality of the magnetic properties of doped cuprates implies a fundamental role that spin fluctuations play in the physics, and a complete understanding of the magnetism has become even more important.

Many cuprates have a tendency to develop stripe-like spin correlations. In doped La_2CuO_4, when the doping is below optimal for the superconductivity, the stripe-like correlations become static, forming incommensurate spin-density waves.[19] For optimal and higher doping the static order disappears, but dynamic correlations persist up to the superconductor-normal-metal boundary.[20] The disappearance of the stripe-like correlations at the superconductor-normal-metal boundary suggests that dynamic stripes are necessary for the superconductivity, as some theories have predicted.[21] However, *static* magnetic order clearly competes with the superconducting order.[19] The nature of this competition has been studied by recent magnetic field experiments on La_2CuO_{4+y},[22,23] and $La_{2-x}Sr_xCuO_4$.[24,25] These experiments show that the magnetic moment associated with static long-range SDW order increases when the superconductivity is weakened by the applied magnetic field. Note that in electron-doped cuprate superconductors, like $Nd_{2-x}Ce_xCuO_4$ and $Pr_{1-x}LaCe_xCuO_4$, static magnetism also coexists with the superconductivity. In this case too, it appears

that the ordered magnetic moment increases with applied magnetic field,[10,26] but it is not clear whether the mechanism responsible for this increase is the same as in the hole-doped cuprates.[27]

Magnetic correlations in $La_{2-x}Sr_xCuO_4$ reveal themselves in neutron scattering measurements as a quartet of incommensurate peaks around the antiferromagnetic zone center (at reciprocal lattice positions $(1 \pm \delta, \pm\delta)$ in orthorhombic notation, where $\delta \simeq 0.125$ for $x \geq 0.125$). In $La_{2-x}Sr_xCuO_4$, previous studies have shown that static magnetic order exists for $x \leq 0.13$, while for larger x there are gapped spin excitations at the same positions in reciprocal space; gap values of 4 to 8 meV are measured for $x \geq 0.15$ depending on the doping level.[28,29] Previous neutron scattering experiments in magnetic fields have revealed either an enhancement of the static SDW order in underdoped $La_{2-x}Sr_xCuO_4$ and La_2CuO_{4+y} superconductors[22,23,24,25] or changes in the dynamic spin susceptibility in optimally doped or slightly overdoped $La_{2-x}Sr_xCuO_4$.[11,30,31] In the former case, the intensity of the magnetic Bragg peaks increases with magnetic field accompanying the suppression of the superconducting order parameter. In the latter case, the magnetic field enhances the spin susceptibility at energies below the gap.

As a result of these experiments, a theoretical phase diagram of doped La_2CuO_4 superconductors has been proposed for T = 0 K.[32,33] Fig. 1a shows a fragment of this phase diagram, adopted from Refs. 4,32. The horizontal axis r is a measure of the repulsive coupling between the superconducting and magnetic order parameters; it is assumed to be approximately proportional to doping. The vertical axis is the magnetic field. There exist three distinct phases: (i) the spin-density-wave (SDW) phase at high field, presumably above the upper critical field, where superconductivity is destroyed by the field, (ii) the superconducting (SC) phase at high doping and small fields, and (iii) the intermediate "SC+SDW" phase, where the SC and SDW order parameters coexist. The boundary between the SC and SC+SDW phases is a line of quantum phase transitions. The left vertical arrow shows the approximate trajectory through the phase diagram for the earlier experiments on underdoped $La_{2-x}Sr_xCuO_4$ and La_2CuO_{4+y}.[22,23,24,25] These materials are in the phase where the magnetic and superconducting order parameters co-exist and compete microscopically. This phase is transformed into a SC magnetically disordered phase with increased doping. In the latter phase, the spin susceptibility increases with magnetic field at low energies because the applied field brings the system closer to the magnetic ordering transition while suppressing

the superconducting order. The phase diagram predicts that the spin-ordering transition can be achieved either by reducing the doping or applying a strong enough magnetic field. However, substitutional doping affects both the superconducting and magnetic order parameters, as well as the degree of disorder in the samples. In this sense, achieving the phase transition by applying a magnetic field would be the most straightforward measurement of the phase transition.

The experiments presented here demonstrate directly that the spin-ordering transition can be induced by a magnetic field in slightly underdoped $La_{2-x}Sr_xCuO_4$ with nominal $x = 0.144$. Fig. 1b shows the field dependence of one of the incommensurate magnetic Bragg peaks for this material. At zero applied field, no elastic signal is found, as expected for this doping level. Static long-range magnetic order appears above approximately 3 T. This result is direct evidence of the quantum phase transition between magnetically-ordered and disordered superconducting states in $La_{2-x}Sr_xCuO_4$.

The single crystal of $La_{2-x}Sr_xCuO_4$ with nominal Sr concentration $x = 0.144$ was grown by the travelling solvent floating zone technique with subsequent annealing in oxygen atmosphere at 900 °C for 24 hours followed by furnace cooling in oxygen. We estimate the present sample doping $x = 0.144 \pm 0.005$. The preparation conditions are essentially identical to other samples of $La_{2-x}Sr_xCuO_4$ ($x = 0.1$, 0.12, 0.13, 0.15, etc.) used to study the SDW order and fluctuations (see refs. 24,28 and refs. therein), which assures consistency in Sr and oxygen contents. Magnetic susceptibility of a small piece cut from the neutron scattering sample showed a superconducting transition with T_c (onset) = 37 K, centered at 35 K. The sample of 8 g was cut into two equal pieces, which were co-mounted in order to fit inside the split-coil 14.5 T magnet; the mosaic of the assembly was 0.5° (FWHM). Elastic neutron scattering experiments in the presence of a high magnetic field were performed at the Hahn-Meitner Institute, utilizing the FLEX cold-neutron triple-axis spectrometer with incident neutron energy of 4 meV.

Fig. 1b shows the field dependence of the intensity of one of the SDW peaks in the $x = 0.144$ sample. Similar behavior is found for the other incommensurate peaks in this sample. The intensity is fitted according to a power law $I(H) = I_0 + A(H - H_c)^{2\beta}$, as expected near a second-order phase transition. Here I_0 is a constant background, $H_c = (2.7 \pm 0.8)$ T and $\beta = (0.36 \pm 0.10)$ are fitting parameters. The fit is shown as a solid curve in Fig. 1b. The current statistical error bars do not allow us to draw conclusions about the universality

class of the transition.

Fig. 2 shows three out of four incommensurate SDW peaks. Comparing the scans at $H = 0$ T for $T = 40$ K and 1.5 K shows that the scans in zero field do not change above and below the superconducting T_c within the errors. The absence of a peak places an upper bound on the zero-field ordered magnetic moment of $0.02\mu_B$, assuming a resolution-limited correlation length. The peaks in Fig. 2 B and C are resolution limited at 14.5 T, which places a lower limit on the magnetic correlation length of $120\AA$ (the data in Fig. 2 A is too noisy to allow for a reliable fit). The ordered moment at 14.5 T is $(0.06 \pm 0.02)\mu_B$.[34] Fig. 2B also shows the peak at $H = 7$ T, where the intensity is approximately half of that at $H = 14.5$ T.

The behavior of the $x = 0.144$ sample is qualitatively and quantitatively different from that of previously studied samples of $La_{2-x}Sr_xCuO_4$ ($x = 0.1$ and 0.12) and La_2CuO_{4+y}, where an enhancement of an *existing* static SDW order was found.[22,23,24,25] As mentioned above, the scans in Fig. 2 suggest that for $x = 0.144$ the magnetic moment is zero when $H = 0$ T. In addition, the field dependence shown in Fig. 1b is consistent with the absence of SDW order below ~ 2.7 T. In the previous results for $La_{1.9}Sr_{0.1}CuO_4$, the field-induced signal at 2.5 Tesla is about 35% of the signal induced at 14 T.[25] If a comparable increase existed for our sample, the signal at 2.5 Tesla should be ~ 1280 counts/20 min. As seen in Fig. 1b, such a signal is not observed and would be clearly outside of the error bars. In addition, the field dependence for $La_{1.9}Sr_{0.1}CuO_4$ is fit by a line which has an infinite slope in limit of $H \to 0$. Again, our data do not support this and are qualitatively different. The doping of $x = 0.144$ was chosen to be close to $x = 0.15$ for which it is known that the ground state has complete spin-gap.

It is useful now to refer to the phase diagram in the Fig. 1a. The left vertical arrow corresponds to the $x = 0.10, 0.12$ and La_2CuO_{4+y} samples, which are in the "SC+SDW" phase at zero field.[22,23,24,25] By contrast, the $x = 0.144$ sample is in the SC phase at $H = 0$ T and crosses into the SC+SDW phase as the field increases. This behavior corresponds to the right vertical arrow in the phase diagram. Note that the high-field SDW state is an actual thermodynamic phase since we observe long-range SDW order above the transition. Given that a static, long-range ordered SDW phase is found at $H = 0$ in $La_{2-x}Sr_xCuO_4$ for $x = 0.12$,[35] but not in our sample with $x = 0.144 \pm 0.005$, we conclude that the quantum critical point at $H = 0$ T lies between $x = 0.12$ and $x = 0.14$. We will discuss this further

below.

Most models for the enhancement of SDW order or fluctuations by the magnetic field assume a competition between the superconductivity and magnetic order, although different models for this competition have been proposed.[32,33,36,37,38] Our experimental results, combined with the earlier experiments[22,23,24,25] favor the model proposed in Ref. 32. This model assumes a microscopic competition between SDW and SC order. This is a Ginzburg-Landau model with a repulsive coupling between the superconducting and magnetic order parameters near the magnetic ordering phase transition. The magnetic field penetrates type-II superconductors in the form of magnetic flux lines and the superconducting order parameter is suppressed to zero inside the vortex cores and recovers to its zero-field value over a large length scale away from the cores. This suppression of the superconducting order parameter leads to an enhancement in the competing magnetic order far away from the vortex cores and ensures that the magnetic correlation length spans many inter-vortex distances.

This leads us to propose a possible scenario for the field-induced transition. It is well known that in the SC phase the spins fluctuate at various frequencies, which may vary depending on local disorder or doping variations. These 2-dimensional spin fluctuations at $Q = (1 \pm \delta, \pm\delta)$ around the antiferromagnetic zone center are well characterized by inelastic neutron scattering.[1,39] When a magnetic field is applied, the superconductivity is completely suppressed within the vortex cores, and partially suppressed even at large distances from the vortices. Because of the competition between SC and SDW order the magnetic fluctuations are enhanced where SC is suppressed, and their spectral weight distribution is modified such that lower frequency fluctuations become stronger.[11,30,31] When the field exceeds a critical value, the regions of slowest spin fluctuations become large enough that long-range magnetic order can develop in the form of static incommensurate SDW order at the same $Q = (1 \pm \delta, \pm\delta)$. When the field is increased further, the magnetic order parameter is stabilized at the expense of the superconducting order. It is expected that away from the critical regime near $H_c(x)$ (and well below H_{c2} where the superconductivity is completely destroyed), the elastic neutron scattering intensity should follow an $(H/H_{c2})\ln(H_{c2}/H)$ dependence,[40] similar to the samples where the SDW order exists at zero field.[22,23,25] However, our experiments are close enough to the phase transition that they are dominated by the critical fluctuations. The model of Ref. 32 predicts a linear behavior of the intensity with the field, which is not inconsistent with the data of Fig. 1b.[40]

In the $La_{2-x}Sr_xCuO_4$ phase diagram, static magnetic correlations are observed for $x \leq 0.13$. However, the static correlation length is short-ranged for $x = 0.13$ ($\xi \simeq 88\mathring{A}$),[41] whereas it diverges for $x \simeq 0.12$ ($\xi > 200\mathring{A}$).[35] Therefore, the $H = 0$ quantum critical point separating the SC+SDW and SC phases is close to $x = 0.125$. The short-range SDW peaks observed for larger x would then correspond to critical scattering. Indeed, for a sample with $x = 0.14$, we have observed short-range SDW order in zero-field and a transition to long-range order with increasing field,[42] consistent with this picture. For concentrations near but below $x = 0.125$, the SDW peaks are resolution-limited, and long-range order is observed. However, as x is decreased further, the static correlation length at low temperatures is again measurable and finite.[43] This behavior is consistent with an incommensurate system with structural (i.e., random field) disorder. As the system becomes more incommensurate, the random fields become more important. The QCP appears near $x = 0.125$ because that is the concentration at which the system is closest to being commensurate. Note that random fields are much less important for commensurate systems. Similar behavior has been recently discussed in relation to an avoided critical point on the phase diagram by Kivelson and coworkers.[33]

Finally, we note a possible connection between the present results and the spin gap phenomenon, which is observed in $La_{2-x}Sr_xCuO_4$ for $x \geq 0.15$.[29,44] One theoretical model associates the spin gap with the proximity to a quantum phase transition between the superconducting phase with no magnetic order and a SC+SDW phase, where SC and SDW order parameters coexist.[3] The spin fluctuations in the disordered phase are expected to be gapped near a transition to the magnetically-ordered phase because of the critical slowing down of the quantum fluctuations. The gap energy is $E_{gap} \sim \hbar/\tau$, where τ is the characteristic time scale of spin fluctuations. This behavior is in contrast to a classical phase transition, where the Heisenberg uncertainty relation does not apply and the spin-gap is not expected to form in the disordered phase. Earlier indirect evidence for a presence of quantum phase transition has come from experiments on $La_{2-x}Sr_xCuO_4$, $x \simeq 0.14$, which showed a diverging correlation length of the spin fluctuations, but do not identify the two phases that are separated by this transition.[39] Inelastic neutron scattering measurements on our $x = 0.144$ sample would be of great interest to explore the relationship between the spin-gap in $La_{2-x}Sr_xCuO_4$ and the spin-ordering phase transition reported here.

In conclusion, we have observed that in slightly underdoped $La_{2-x}Sr_xCuO_4$ there is a

magnetic-field-induced transition between the magnetically disordered state and the magnetically order state with incommensurate SDW order coexisting with the superconductivity. In addition, since long-range SDW order can be induced by an applied field, it appears that the SDW phase is not the result of chemical inhomogeneities, but rather is a true thermodynamic phase. Our results are consistent with a picture in which the $H = 0$ quantum-critical point lies close to the doping level of $x = 0.125$. Further experiments, especially measurements of the instantaneous correlation length by neutron scattering, would be most interesting to study the critical behavior near this point.

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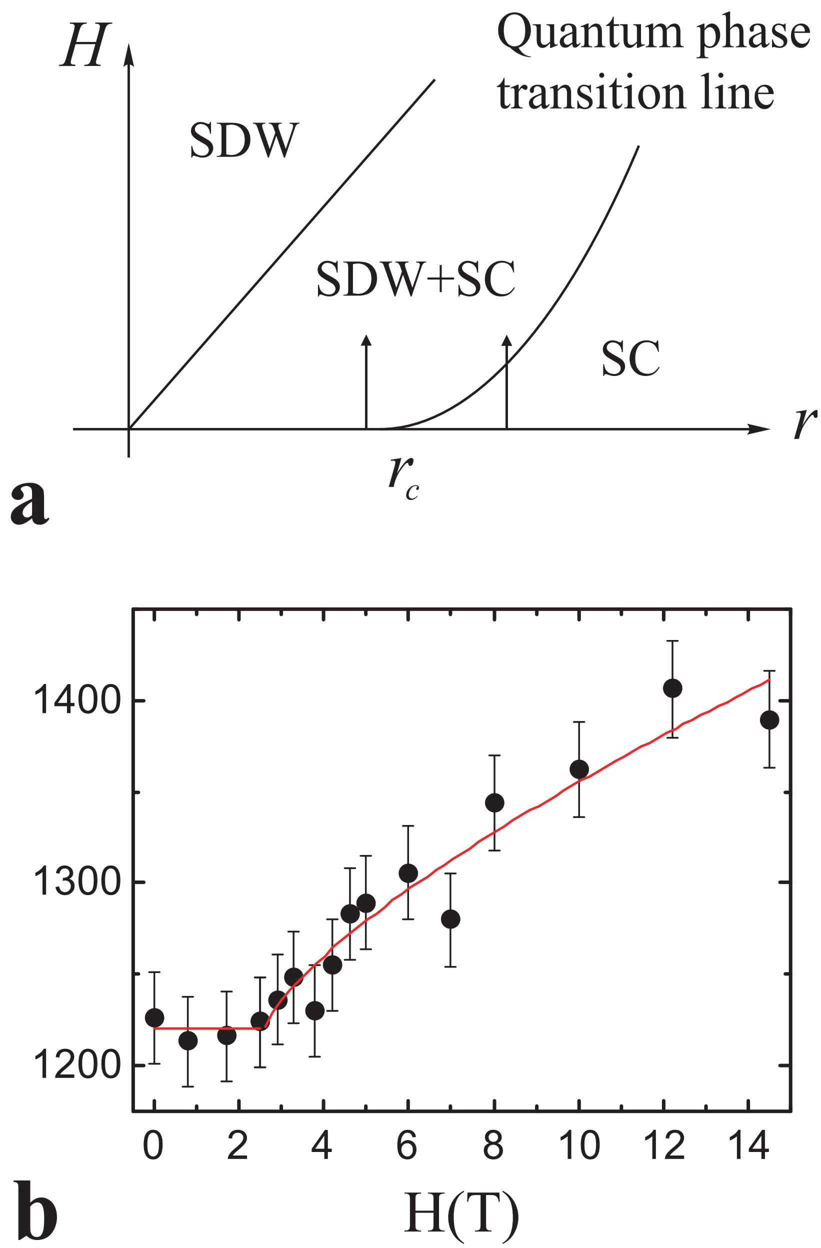

FIG. 1: (a) A fragment of the theoretical phase diagram, adopted from Refs. 4,32. The vertical axis is the magnetic field and the horizontal axis is the coupling strength between superconductivity and magnetic order. (b) Field dependence of the magnetic Bragg peak corresponding to the incommensurate SDW peak at Q = (1.125,0.125,0). Every point is measured after field cooling at T = 1.5 K. The data are fitted to $I = I_0 + A|H - H_c|^{2\beta}$ above H_c as explained in the text. Spectrometer configuration: 45-60-Be-S-Be-60-open; cold Be filters were used before and after the sample to eliminate contamination from high energy neutrons; E = 4 meV.

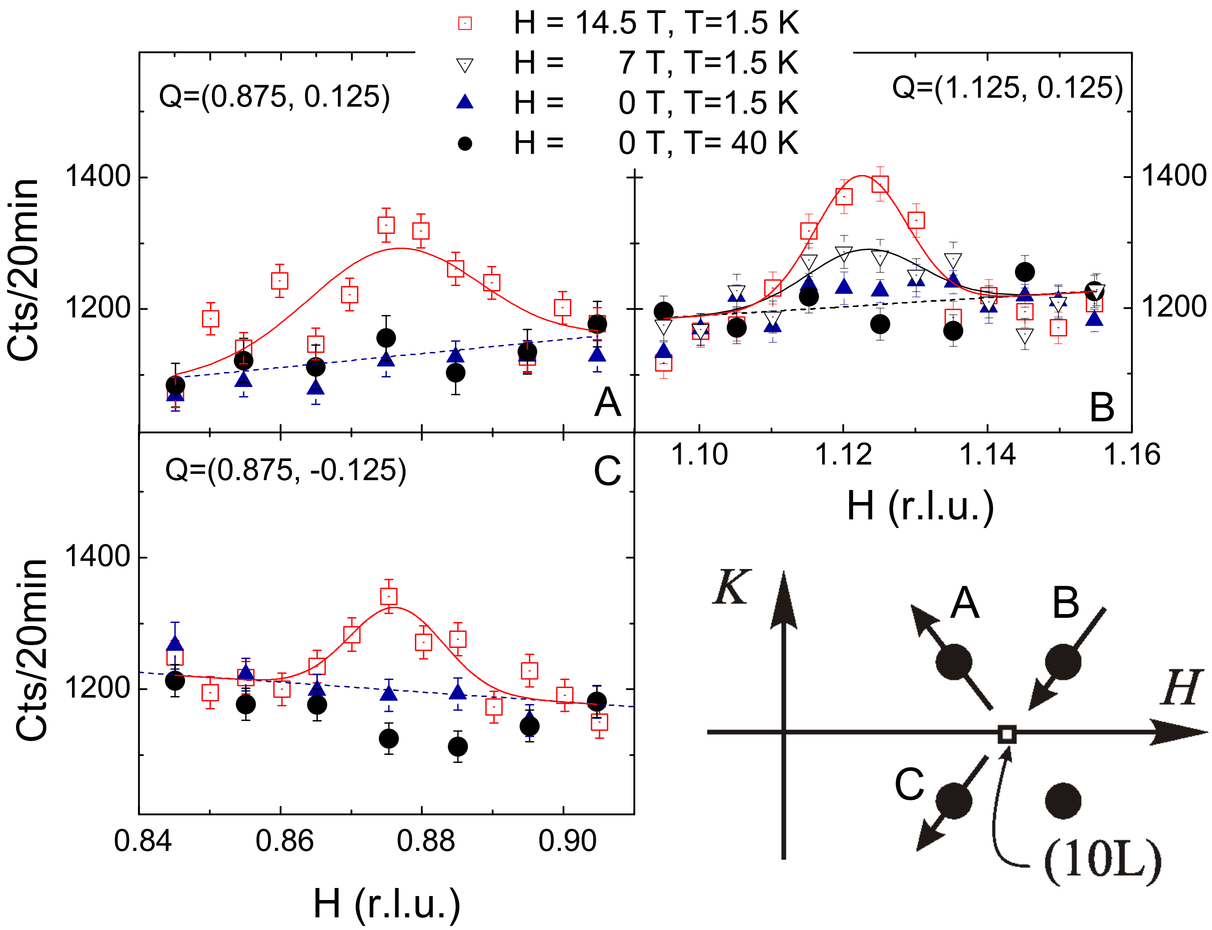

FIG. 2: Incommensurate SDW peaks in the $x = 0.144$ sample. The peaks are shown at three different positions around the AFM zone center at different magnetic fields for $T = 1.5$ K and for $H = 0$ T at 40 K, which is above $T_c = 37$ K and above the highest temperature at which the SDW order is formed in $La_{2-x}Sr_xCuO_4$. The peak positions and scans directions are shown schematically in the inset. Magnetic field scans were performed after field-cooling. The zero-field background data are fitted by a linear function and the field-induced magnetic Bragg peaks are fitted by a Gaussian above this linear background.